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A LATEX IPN COATING SYSTEM FOR DAMPING NOISE AND VIBRATIONS OVER A BROAD TEMPERATURE RANGE

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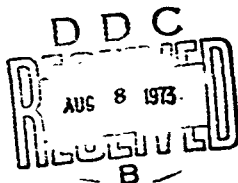
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A Latex IPN Coating System for Damping

Noise and Vibrations over a Broad Temperature

Range

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Abstract

Based on the principles of constrained layer damping, "Silent Paint" is a two-layer coating system capable of attenuating noise and vibration over a broad temperature range. The damping undercoat layer is composed primarily of a methacrylic /acrylic semi-compatible interpenetrating polymer network in latex form, with thickeners and preservative added. The constraining overcoat layer is based on a reinforced epoxy resin, and exhibits great stiffness. The total system, exhibiting significant damping behavior from $-30^{\circ}C$. to over $+70^{\circ}C$., used in the form of coatings put easy application on arbitrary substrate configurations. The synthesis of the latex IPN is summarized, and the mechanical and damping behavior of the new coating system is compared to several materials intended for use as noise and vibration damping products.

Introduction

The need to attenuate unwanted noise has become a problem of major proportions in the technologically advanced nations.[1,2] During the past several years, several scientific and engineering papers were published, largely describing extensional damping materials,[5-8] and several patents of importance were issued.[9-12] Such materials are often applied by trowling techniques to form a thick mastic layer. While single layer, extensional damping materials are more easily applied, the greater efficiency of constrained layer damping techniques has long been recognized,[8,9,12,13-15] wherein a damping layer is sandwiched between the vibrating substrate and a stiff outer layer.

The most common types of damping materials employed involve simple homopolymers or copolymers, with efficient damping limited to a temperature range of approximately 20-30°C., centered about the glass-rubber transition of the polymer involved.[16-18] Incompatible polymer blends and grafts[19-21] with widely separated glass-rubber transition zones, exhibit two such damping ranges with little damping in between. Semi-compatible polymer blends and grafts,[22,23] however, where the mixing of the two kinds of polymer molecules is extensive but incomplete, lead to a broader temperature use range.

One way of attaining intimate mixing is through the use of interpenetrating polymer networks (IPN's)[24-27], which are a unique type of polymer blend, synthesized by swelling a cross-linked polymer (I) with a second monomer (II), plus crosslinking and activating agents, and polymerizing monomer II in situ. The term IPN was adopted because, in the limiting case of high compatibility

between crosslinked polymers I and II, both networks can be
visualized as being interpenetrating and continuous. However, if
components I and II consist of chemically distinct polymers, in-
compatibility and some degree of phase separation usually result,
the exact extent depending upon the mixing thermodynamics[26,27]
and crosslink density[28] of I and II. Under conditions favorable
to damping behavior, phase domains of the order of 100 $\overset{\circ}{A}$ develop.[25]
For ease of application as coatings, IPN's in latex form,[26,27]
rather than bulk form[24,25,28] will be considered.

Silent Paint

The purpose of the present initial engineering study was to
explore the feasibility of developing a latex IPN "Silent Paint",
as the material was nicknamed in our laboratory. A constrained
layer configuration was adopted, as illustrated in Figure 1. The
thin panel substrate A is first coated with a damping material, B.
Ideally, B should have anti-oxidant and preservative properties
normally incorporated in undercoat materials as well as damping
properties. Since a relatively thick layer (5 to 20 mils) was
envisioned, a high viscosity for the material was also desired to
reduce sagging. After suitable drying times, the constraining
layer C would be applied. While the most important feature of C
involved great stiffness after drying (i.e., a very high modulus),
a material that would afford reasonable abrasion and mechanical
protection was also desired. In particular, three technical
objectives for the new damping system were sought:

(1) To develop a material with a broad temperature damping

range, with emphasis on the span between -20°C. and $+50^{\circ}$C. The acoustical span of frequencies, 20 Hz to 20,000 Hz, should be covered.

(2) To develop a material wherein both the damping layer and the constraining layer could be applied in the form of coatings, by spraying, dipping, or brushing. Thus a wide range of substrate configurations could be easily damped.

(3) To develop a material which would have the protective properties of coatings, i.e., against corrosion and mechanical abrasion, and indeed be the choice of coating where noise attenuation was also desired. Application in the present case would be limited to thin panels, such as air conditioner and machinery housings, or automotive bodies; or lockers and cabinets. This limitations arises because the damping layer thickness must be an appreciable fraction of the substrate thickness.[13,15] Also, the coated substrate portions should be the primary source of the noise or vibration, i.e., resonant vibration.

Experimental

Materials

The preparation of the latex IPN materials has been previously described.[26,27] In brief, a crosslinked seed latex of poly(echyl methacrylate), PEMA, was prepared. This seed latex was then swollen with butyl acrylate monomer plus crosslinker, and polymerized _in situ_ to form poly(n-butyl acrylate), PnBA. The composition chosen for most of the work reported herein was 25/75 PEMA/PnBA, each network containing 0.4% crosslinker.

The finished latex was thickened with a 50/50 poly (n-butyl acrylate-co-ammonium acrylate). Several minor ingredients were also added to impart anti-oxidant and mildew resistant properties, followed by ammoniation to pH = 8-9. No fillers were added to this damping material. This material was employed as the damping layer.

The constraining layer employed HS7130 epoxy resin (High Strength Plastics, Chicago, Ill., with 5% by weight of Fybex fibers (duPont, Wilmington, Delaware) added to impart a higher modulus. The damping system consisting of a composite of these two layers bears the designation D'. The designation D represents an earlier material having no added fibers in the constraining layer.

Several commercial materials were also studied, used as received. These included extensional damping compounds based on poly(vinyl acetate), F epoxy resin, E, a damping tape with an aluminum backing. Y, an auto undercoating material, G, a latex paint designed for application to bare metal, H, and an acrylic latex flat house paint, S.

Instrumentation

Four groups of experiments were conducted to estimate the mechanical and damping behavior of the individual components of the "Silent Paint", and also the complete composite system. Briefly, these groups include:

(1) A Rheovibron direct reading viscoelastometer, model DDV-II (Vibron, manufactured by the Toyo Measuring Instrument Co., Ltd., Tokyo, Japan.) This instrument deforms a strip of

material in the extensional mode, a frequency of 110 Hz being employed. Studies were conducted as a function of temperature, the temperature range of -130°C. to $+150^{\circ}$C. being covered with a heating rate of about 1°C. per minute.

Vibron studies yield the complex modulus, E^{*}, and loss tangent, $\tan \delta$. The quantity $\tan \delta$ is a direct measure of a material's ability to convert mechanical energy into heat. The storage and loss moduli, E' and E'', respectively, were calculated from the relations

$$E' = E^{*} \cos \delta \qquad (1)$$

$$E'' = E^{*} \sin \delta \qquad (2)$$

(2) A vibrating reed apparatus, employing Vibration Test Equipment model SD-A (the MB manufacturing Co., New Haven, Conn.), was driven by a Hewlett-Packard Model 2020 Low Frequency Oscillator. The motion was detected by an accelerometer (Model 111, Wilcoxen Research, Bethesda, Md.) placed on one end of the reed. The reed itself was fastened to the vibrator midpoint. The assembly was placed in an environmental chamber to allow the damping to be studied over the range of -30°C. to $+70^{\circ}$C. Following initial cooling with liquid nitrogen, the chamber was warmed at about 1°C. per minute, with readings taken every 3-5 minutes. The quantity A obtained from this equipment indicates a relative measure of the amplitude of the damped reed under a fixed driving force, and A_{o} represents the value for the uncoated reed.

The quantity

$$\frac{1}{A} - \frac{1}{A_o} \qquad (3)$$

is proportional to tan δ, and serves as a relative measure of the damping attained. The test frequency was about 600 Hz, the exact frequency being varied so that resonance was retained as the temperature was varied.

(3) A Brüel and Kjaer complex modulus apparatus [15] (Brüel and Kjaer Precision Instrument Co., Cleveland, Ohio), with a Beat Frequency Oscillator and Recorder 2305, was kindly made available by Dr. Edward Gladding, Elastomers Division, Experimental Station, du Pon de Nemours and Co., Wilmington, Del. The B & K equipment employs a cantilever-mounted vibrating reed. Damping is determined from the decay of resonant vibrations after the excitation is removed. The results were reported as percent critical damping, $C/C_o \times 100$, where Co is the damping just sufficient to prevent oscillation of a system. For small damping, tan $\delta = 2C/C_o$.

The reeds themselves in experiments (2) and (3) were cut from Nicholson Precision Ground 01 tool steel flat stock, $\frac{1}{2}'' \times 8''$ $\times$ 62.5 mil thick. The various materials of interest were coated uniformly onto one side of the reeds, with total thicknesses varying from 10 to 15 mils. Material D', the major system of interest, was studied as a function of individual layer thickness.

(4) In a separate experiment, $2' \times 3' \times 38$ mil steel panels were coated with several materials of interest to thicknesses of 14 mils. Through the courtesy of Mr. Georges Garinther, these

panels were tested in the anechoic chamber of the Human Engineering Laboratory, Aberdeen Proving Ground, Aberdeen, Md. The test configuration is shown in Figure 2. The panels were suspended by thin wires attached to the panel corners. A B & K ½" microphone was placed behind the panels, and a 1 3/4" diameter plastic ball was positioned to strike the center of the panel. Two different studies were carried out:

(a) The total noise emitted from 16 to 20,000 Hz was recorded as a function of time, using a B & K 235 graphic level recorder. While the total area under the curve was of some interest, the slope of the curve, called the decay rate, Δ yielded, a reliable measure of noise attenuation. The slope was determined at a convenient level of 15 dB, since Δ varied somewhat depending on the exact portion of the curve considered.

(b) The maximum sound pressure was recorded by 1/3 octave band widths over the audible range. While somewhat limited by the response time of the equipment, this test allowed an analysis of the noise level emitted immediately after the ball struck the panel.

Results

Preliminary Experiments

The slection of 25/75 PEMA/PnBA composition for the latex IPN damping layer was based on additional experiments of the type already reported.[26,27] As shown in Figure 3, the material has nearly constant tan δ from -20°C. to $+50^{\circ}$C. After addition of thickening and preservative agents, the damping characteristics were substantially the same. However, since the usual paint pigments

and fillers detracted from the damping, no fillers were used in the final formulation of the IPN damping layer.

With the constraining layer, on the other hand, the more filler the more effective the material was predicted to be, since stiffness is a major criterion. Epon 828 (Shell Chemical Co.) and HS7130 were filled with systematically increasing quantities of Fybex and glass flock (High Strength Plastics.) The shear modulus times 3 (nearly equal to Young's modulus) of the filled and unfilled materials is shown in table I. The apparent lowering of the modulus at high filler levels was caused by entrapped void and/or improper filler dispersion. For the immediate purpose of attaining materials that were suitable for coatings, HS7130 was selected as the constraining layer for material D. Later, 5% Fybex was mixed with the HS7130 to attain greater stiffness. This latter is approximately three times as stiff as an unfilled epoxy material.

The use of this epoxy also has secondary advantages also, as hown in Figure 4. A small loss peak exists at -30°C. and a broad major loss peak is centered at $+95^{o}$C., the latter representing the glass-rubber transition of the epoxy. Fortuitously, these transitions occur just below and just above the transition and maximum damping range of the latex IPN material. The two-layer damping system is thus capable of significant extensional damping at temperatures outside the original objective of -20 to $+50^{o}$C., as shown later in Figure 7.

Panel Experiments

Figure 5 compares the initial response of the bare steel panel (J) with the first Silent Paint system (D). The maximum sound pressure near 250 Hz in both cases is similar, as expected, but at higher frequencies the sound pressure of the coated panel was less. A possible cause involves the more rapid damping of the high frequency components in relation to the instrument response time. These results are expected on the basis of a constant amount of the mechanical energy being converted to heat per cycle, nearly independent of the frequency. To the ear, the coated panel sounded much deader, when struck.

Figure 6 compares the attenuation of the noise level after the initial excitation, at room temperature, for the same panels. The results of these experiments are summarized in Table II. Panel D has a larger decay rate and a smaller area under the curve than Panels A, B, or C (IPN damping layers with various filler levels), confirming that large quantities of filler in the damping portion are detrimental. Panel D is superior in performance to Panels G and H, roughly equal to F, and somewhat inferior to E.

Vibrating Reed Experiments

Figure 7 compares D', Silent Paint with the Fybex-stiffened constraining layer, with three other materials. Surprisingly, the epoxy material E appears to be most effective at temperatures below and above ambient. This resembles the behavior of the HS7130 epoxy presented earlier in Figure 4, although the chemical compositions of the two epoxies undoubtedly differ somewhat. The poly(vinyl acetate) based material F has its peak just above room temperature, but rapidly loses effectiveness at higher or lower temperatures.

While the damping efficiency of composition D' never quite rises higher than the peaks attained by compositions E and F, most importantly D' remains effective over the entire temperature range of -30°C. to +70°C. The sharp rise at high temperatures is caused by the onset of the epoxy glass-rubber transition, and like damping material E, probably reaches a maximum damping efficiency level in the range 90-100°C. The commercial acrylic latex paint, S, by comparison, has low damping everywhere except in the vicinity of room temperature, where the glass transition of this highly filled material occurs. The low damping is not surprising, inasmuch, as the polymer serves mainly as a binder. Of course, the paint is intended as a protective coating and not as a damping material.*

In an attempt to evaluate the relationship between the damping and constraining layers, the relative thicknesses were systematically varied, holding the total thickness constant. Figure 8 illustrates the relative damping at 25°C. vs thickness ratio. Most strikingly, maximum damping is achieved with a damping/constraining ratio near 7/4. Thus, optimum performance for this system demands that the damping layer be nearly twice as thick as the constraining layer.

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*Comparison of S and D' in Figure 7 resolved a bothersome problem. The writers prepared several sets of demonstration tea bells with S and D' along with other materials, only to discover that D' was only marginally better than S! After placing the bells in a refrigerator, the superior qualities of D' became more apparent.

Complex Modulus Experiments

The first harmonic, with a frequency of approximately 180 Hz was selected for this experiment. The results, expressed as the percent critical damping, $C/C_o \times 100$, in Table III are based on the thin layer approximation. All the materials except Y were 11 mils thick, Y being 13 mils thick. Material D' had 5 mils of damping material and 6 mils of constraining layer. These room temperature results do not, of course, reflect the importance of temperature dependence. Nonetheless, material D', with 1.0% critical damping, may be seen to rank fairly well by this test.

Discussion

Many noise-emitting, vibrating structures are commonly exposed to wide temperature variations. Two examples include outdoor applications, and use in connection with motors, transformers, or refrigeration equipment. "Silent Paint" is designed for ease of application over the arbitrary configurations such sources impose, and indeed may partly replace the normal protective or decorative coatings normally used on most such surfaces.

In the previous section, we have described the preparation and damping behavior of a two-coat noise reducing paint-like material. It should be emphasized that while the damping latex was designed to have nearly constant tan δ values over the temperature range of -20°C. to $+50^{\circ}$C., the useful temperature span is increased by the fortuitous presence of transitions in the epoxy constraining layer immediately on either side of this range. Thus, "Silent Paint" has significant damping capabilities from about -30°C. to $+90^{\circ}$C. By

contrast, simple homopolymers and random copolymers usually damp over a span of 20-30°C., in the vicinity of their glass-rubber transition. While the damping shown in Figure 7, Table II, and Table III indicate that the present materials (D and D') are not superior at any particular temperature, they have a broad temperature range not heretofore available. Materials D and D' should be considered as experimental prototypes; research is in progress to improve and optimize them.

The temperature range covered by the latex component obviously depends upon its chemistry.[26,27] In particular, lower members of the methacrylate/acrylate homologous series exhibit greater effectiveness at elevated temperatures, while higher members are more useful at lower temperatures. The 25/75 PEMA/PnBA composition selected in the present study was designed to cover the more critical range of human need. If the structure to be damped were to be used at only one temperature, or over a very narrow range of temperatures, a homopolymer with an appropriate glass temperature, together with the constraining layer, would be more effecti.e. For use over somewhat broader temperatures, an appropriate IPN combination might be sought.

A word should be mentioned about frequency ranges. Based on the time-temperature superposition principle, increasing the frequency by one decade will cause an increase in the glass-rubber transition temperature by about 6 or 7°C.[16-18] The acoustical frequency range of 16 Hz to 20,000 Hz, roughly three decades, covers about 18 to 20°C. equivalent range of temperature. This range corresponds almost exactly to the breadth normal associated

with homopolymer transitions. Thus, by correct placement of the
glass-rubber transition, a homopolymer will effectively damp all
acoustical frequencies at one temperature, or one frequency over
a range of about 20°C., but not both. The composition "Silent
Paint", will damp all frequencies over all but the very extremes
of the temperature range -30°C. to $+70^{\circ}$C.

We have mentioned that excessive filler in the damping
layer was detrimental to noise attenuation behavior. However, small
amounts of fillers, especially in the shape of platelets, probably
will serve to enhance the effectiveness of an optimized composi-
tion.[8]

The stiffness of the constraining layer is important, since
damping efficiency is roughly proportional to the modulus of this
layer. It turns out that the Young's modulus of nearly all plastic
materials, including epoxy resins, is close to 3×10^{10} dynes/
cm^2.[17,18] This value can be significantly augmented by addition
of particulate or fibrous fillers, especially the latter.[29-32]
A limitation appears to arise from the quantity of fibrous material
that can be incorporated in a polymer or prepolymer and still re-
tain coating characteristics. Because of simplicity, all materials
in the present work were brushed. Spray techniques are anticipated
to yield superior products because greater amounts of filler may
be incorporated. Also most commercial operations involve baking,[33]
although all films reported herein were cured at room temperature.

Examination of Figure 8 reveals that optimum performance
results when the damping layer is somewhat thicker than the constrain-
ing layer. On a weight basis, epoxies are more expensive than

acrylics suggesting that the optimum overall composition may be a thin coat of very stiff epoxy upon a thick coat of damping material.

In summary, the damping and noise attenuation studies carried out on "Silent Paint" compositions indicate a broad temperature use range, with significant engineering potential. Since research and engineering on damping are continuing in our laboratories, this paper should be considered as a preliminary report.

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Table I

Stiffness Values for Reinforced Epoxys

Materials	$3G(dynes/cm^2)$
Epon 828*	2.0×10^{10}
Epon 828/5% Fybex	2.6×10^{10}
Epon 828/10% Fybex	3.1×10^{10}
Epon 838/15% Fybex	4.0×10^{10}
Epon 828/20% Fybex	3.2×10^{10}**
Epon 828/5% Glass Flock	3.0×10^{10}
Epon 828/10% Glass Flock	3.3×10^{10}
Epon 828/15% Glass Flock	3.1×10^{10}
Epon 828/20% Glass Flock	3.1×10^{10}
HS7130	6.2×10^{10}
HS7130/5% Fybex	9.1×10^{10}
HS7130/10% fybex	6.3×10^{10}
Hs7130/5% Glass Flock	8.0×10^{10}
HS7130/10% Glass Flock	9.0×10^{10}

* The Epon (Shell Chemical Co.) series materials were cured with 12% by weight diethylene triamine.

** General remark: High concentrations of fibers led to voids or air bubbles, artificially reducing the modulus.

Table II

Summary of Steel Panel Damping Experiments

Sample	Δ at 15 dB, $\dfrac{dB}{sec.}$	Total Area, dB $\times$ sec. $\times 10^6$
A	12.4	4.04
B	13.4	4.38
C	12.7	4.0
D	19.5	5.28
E	29.4	2.40
F	17.7	2.65
G	12.0	7.63
H	11.5	5.34
J	8.51	7.76

Table III

Percent Critical Damping at 25°C

Material	$C/C_o \times 100$
J	0.6
S	0.7
H	0.7
E	0.8
F	1.0
D'	1.0
Y	1.3

Figure Captions

Figure 1. Constrained layer damping configuration.

Figure 2. Test configuration for panel damping studies.

Figure 3. Storage modulus, E', loss modulus, E'', and tan δ values for the latex IPN material selected for this study. The broad, controlled glass-rubber transition spanning the range of $-20^{\circ}C.$ to $+50^{\circ}C.$ permits nearly constant tan δ values to be generated.

Figure 4. The storage and loss modulus of the Fybex reinforced epoxy resin, intended for the constraining layer.

Figure 5. Initial sound pressure in dB vs frequency for damping system D compared to the uncoated panel J.

Figure 6. Noise decay of D compared to J, after the steel panels were struck with plastic balls with equal force.

Figure 7. Temperature dependence of damping for D', E, F, and S.

Figure 8. Damping vs composition, holding the total system thickness at 11 mils. Ratios reported as damping/constraining layer thickness in mils.
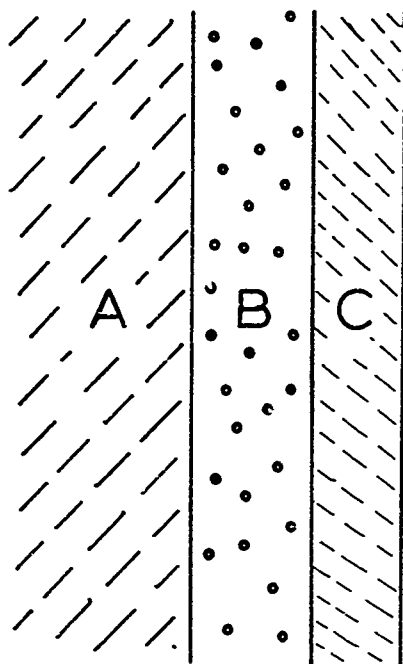
A = Substrate

B = Damping layer

C = Constrain- ing layer

Constrained Layer Damping Configuration
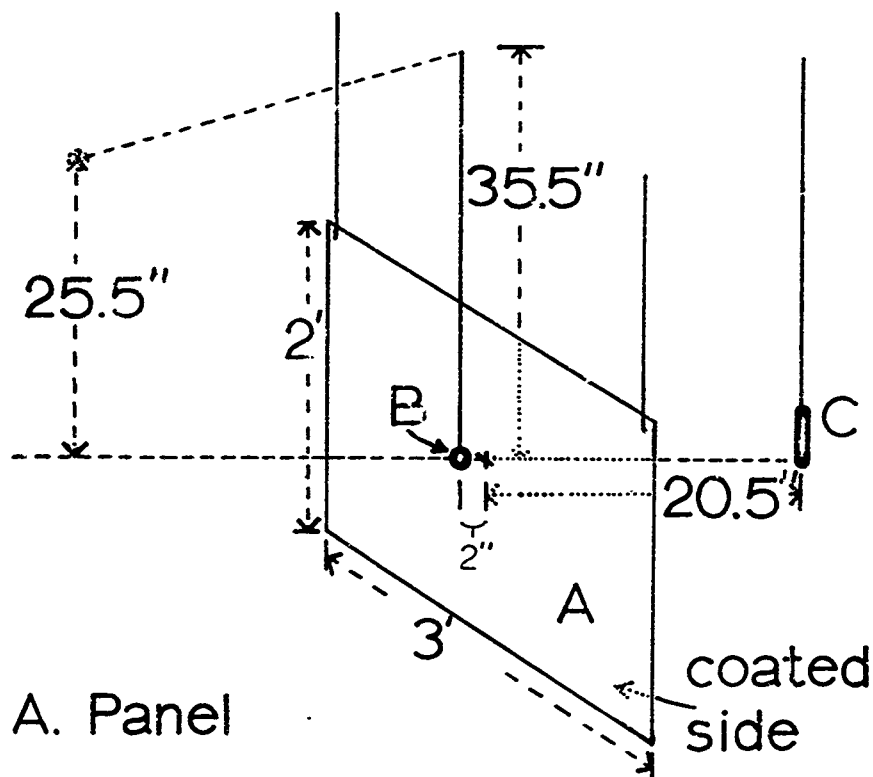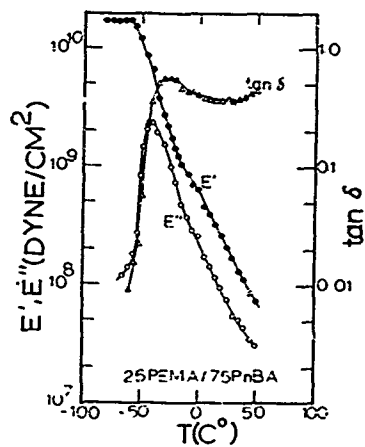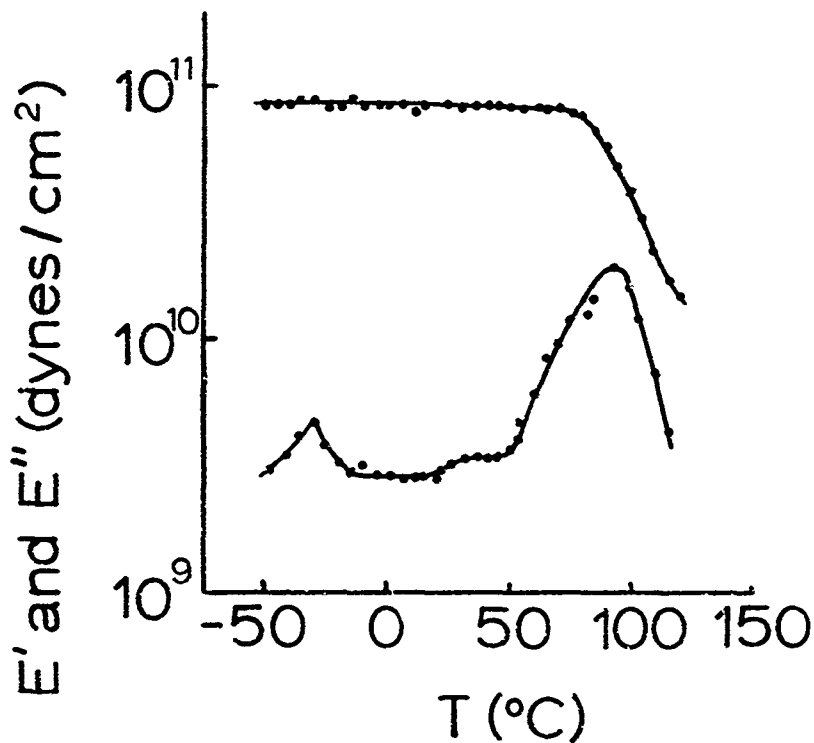
A. Panel

B. Plastic ball

C. Microphone

Figure 5.
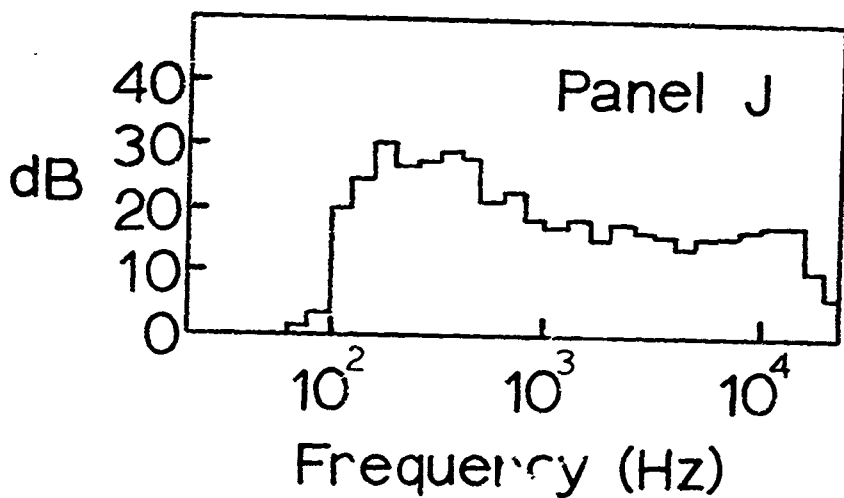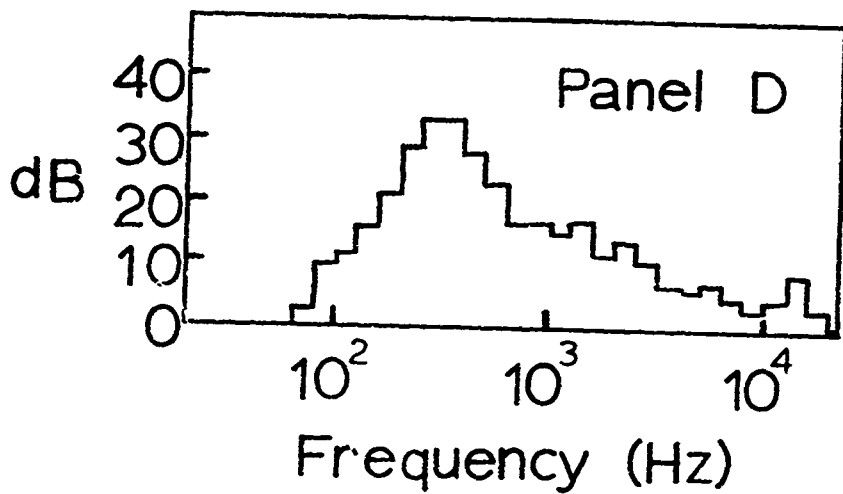
Reinforced Epoxy Coat

Panel D

dB

Frequency (Hz)

Panel J

dB

Frequency (Hz)
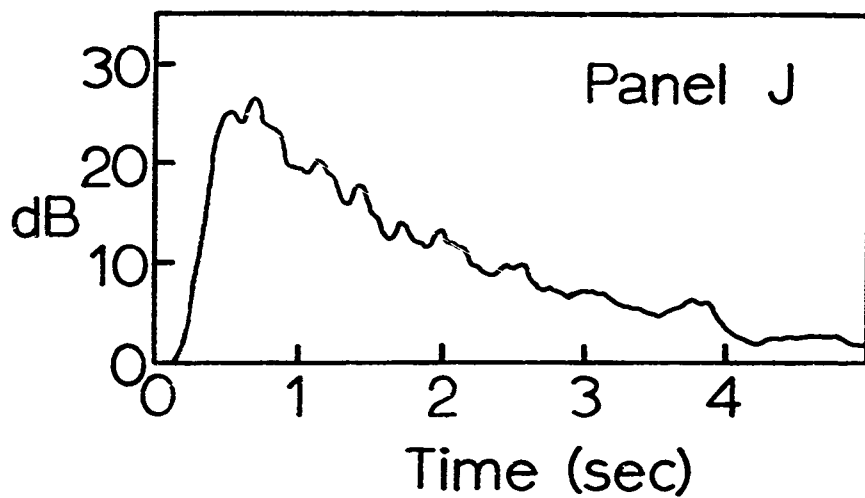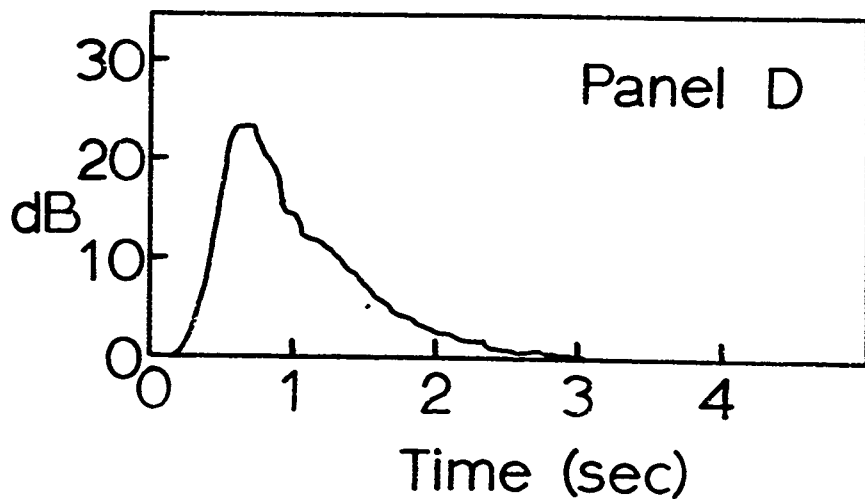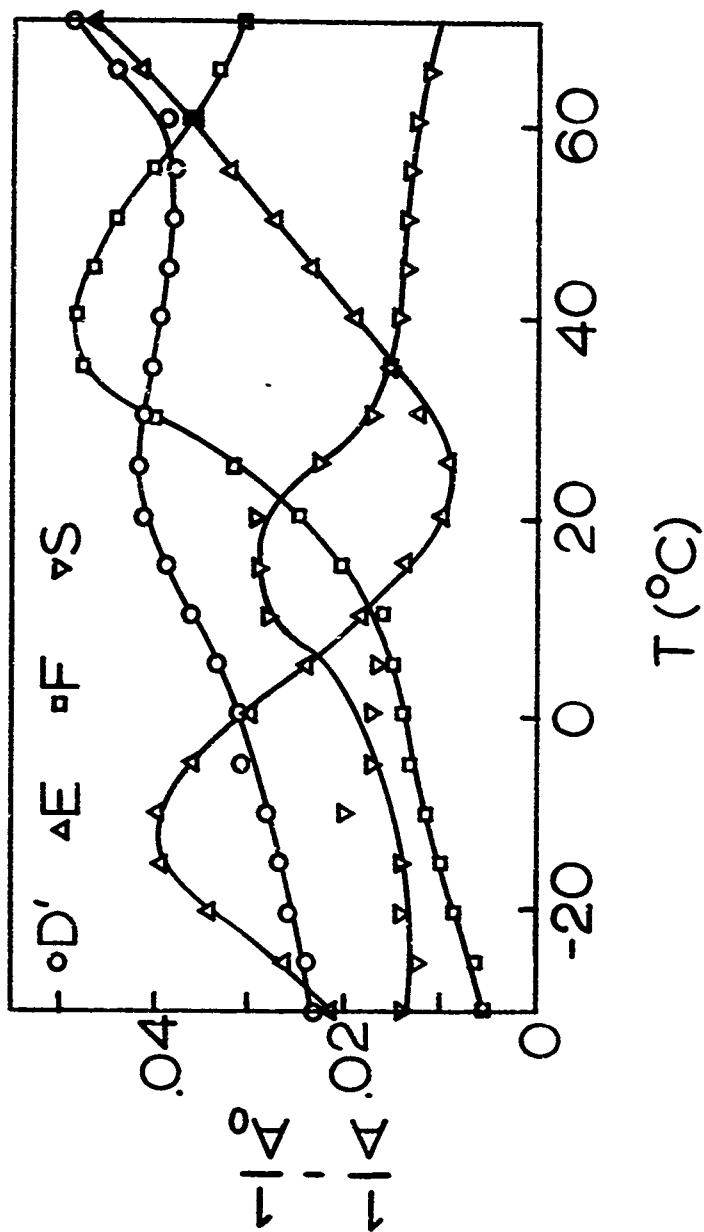
Damping vs. Relative Layer Thickness